

REMARKS

Applicants thank the Examiner for the thorough examination given the present application.

Status of the Claims

Claims 1-10 and 12-16 are pending in the above-identified application. Claims 1 and 10 have been amended. Support for the recitations in claims 1 and 10 can be found in the publication of the present specification, *inter alia*, at paragraph [0026]. Thus, no new matter has been added.

Applicants submit that the present Amendment reduces the number of issues under consideration and places the case in condition for allowance. Alternatively, entry of the present amendment is proper to place the claims in better form for appeal.

In view of the following remarks, Applicants respectfully request that the Examiner withdraw all rejections and allow the currently pending claims.

Drawings

Applicants thank the Examiner for indicating that the drawings are accepted.

Issues under 35 U.S.C. § 103(a)

- 1) Claims 1-6 and 8-9 are rejected under 35 U.S.C. § 103(a) as being unpatentable over Mizumoto et al. '263 (US 4,631,263).
- 2) Claim 7 is rejected under 35 U.S.C. § 103(a) as being unpatentable over Mizumoto et al. '263 in view of Nishino et al. '355 (JP 55-149355).
- 3) Claims 10 and 12-16 are rejected under 35 U.S.C. § 103(a) as being unpatentable over Yokota et al. '063 (US 4,625,063) in view of Mizumoto et al. '263.

Applicants respectfully traverse. Reconsideration and withdrawal of the rejections are respectfully requested based on the following considerations.

Legal Standard for Determining Prima Facie Obviousness

MPEP 2141 sets forth the guidelines in determining obviousness. First, the Examiner has to take into account the factual inquiries set forth in *Graham v. John Deere*, 383 U.S. 1, 17, 148 USPQ 459, 467 (1966), which has provided the controlling framework for an obviousness analysis. The four *Graham* factors are:

- (a) determining the scope and content of the prior art;
- (b) ascertaining the differences between the prior art and the claims in issue;
- (c) resolving the level of ordinary skill in the pertinent art; and
- (d) evaluating any evidence of secondary considerations.

Graham v. John Deere, 383 U.S. 1, 17, 148 USPQ 459, 467 (1966).

Second, the Examiner has to provide some rationale for determining obviousness. MPEP 2143 sets forth some rationales that were established in the recent decision of *KSR International Co. v Teleflex Inc.*, 82 USPQ2d 1385 (U.S. 2007). Exemplary rationales that may support a conclusion of obviousness include:

- (a) combining prior art elements according to known methods to yield predictable results;
- (b) simple substitution of one known element for another to obtain predictable results;
- (c) use of known technique to improve similar devices (methods, or products) in the same way;
- (d) applying a known technique to a known device (method, or product) ready for improvement to yield predictable results;
- (e) “obvious to try” – choosing from a finite number of identified, predictable solutions, with a reasonable expectation of success
- (f) known work in one field of endeavor may prompt variations of it for use in either the same field or a different one based on design incentives or other market forces if the variations are predictable to one of ordinary skill in the art;
- (g) some teaching, suggestion, or motivation in the prior art that would have led one of ordinary skill to modify the prior art reference or to combine prior art reference teachings to arrive at the claimed invention.

As the MPEP directs, all claim limitations must be considered in view of the cited prior art in order to establish a *prima facie* case of obviousness. *See* MPEP 2143.03.

Distinctions over the Cited References

In the outstanding Office Action, the Examiner acknowledges the previous argument that Mizumoto et al. '263 fail to disclose that the interior of the catalyst can be used as sites for the reaction. However, the Examiner asserts that this feature is not recited in the claims. As amended, claim 1 now recites that "the inside of the catalyst is a site of reaction for producing said tertiary amine," and claim 10 now recites "a step of reacting an alcohol with a primary or secondary amine inside a film-type catalyst." The cited references fail to disclose these features.

On page 4 of the outstanding Office Action, the Examiner states, "Such water-repellent catalysts are used to wide range of gas/liquid reactions. Gas/liquid reactions include reactants consisting of gas and liquid and effect a chemical conversion between these reactants (col.2, lines 50-58). It meets the instant claimed limitation of the mass (i.e. gas) transfer between the inside and outside of the catalyst.

In the present specification, the reactants of the amination reaction may be present in a gaseous or liquid phase (paragraph [0060] of the present published application). The process of transferring the reactants and the product in the inside of the catalyst is governed by diffusion, and the distance is reduced to 500 μm or less, whereby the mass transfer between the inside and outside of the catalyst can be promoted thereby effectively utilizing the whole of the catalyst and simultaneously suppressing the excessive reaction of the intermediate reaction product in the inside of the catalyst (paragraph [0026] of the present published application). The diffusivity, which governs the mass transfer of reactants and product in the inside of the catalyst, has been well acknowledged to be much smaller in the liquid state than in the gaseous state, as disclosed in many articles. For example, in the enclosed Bird et al. reference (*Transport Phenomena*), some typical diffusivity coefficients in a gaseous state are disclosed to be of the order of 0.1 cm^2/sec (see page 503, **Table 16.2-2**), while those in a liquid state are of the order of $10^5 \text{ cm}^2/\text{sec}$ (see page 504, **Table 16.2-3**). As such, the film type catalyst of the present invention is much more effective for producing tertiary amine than Mizumoto et al. '263, especially when the reactants and product are in a liquid state.

Moreover, claims 1 and 10 specifically recite that “said film-type catalyst comprises catalyst particles bound to one another via a synthetic resin as a binder” and that “said particles form a three-dimensional network structure via the binder on a substrate.” Accordingly, Mizumoto et al. ‘263 fail to disclose the substrate as recited in the pending claims.

Furthermore, claims 1 and 10 recite “whereby the diffusion rate in the catalyst layer is increased due to said three-dimensional network structure, and the mass transfer between the inside and outside of the catalyst can be promoted thereby utilizing the whole of the catalyst and simultaneously suppressing the excessive reaction of an intermediate reaction product in the inside of the catalyst.”

The Examiner relies on column 2, lines 27-32 of Mizumoto et al. ‘263, which states, “When the water-repellent catalyst of this invention is used, it becomes possible that the gas passes not only over the surfaces of the catalyst but also through the interior of the catalyst and, accordingly, three-phase interfaces are easily formed, and the rate of reaction can be increased.” As noted above, the present invention utilizes the whole of the catalyst. In contrast, Mizumoto et al. ‘263 only uses the interior of the catalyst for permeating the gas. Mizumoto et al. ‘263 fail to disclose that the interior of the catalyst can be used as sites for the reaction. In fact, Mizumoto et al. ‘263 state that the liquid does not penetrate into the catalyst because of liquid impermeability (col. 2, lines 18-19).

The gas/liquid reactions in the presence of a water-repellent catalyst proceed through the formation of three-phase interfaces on the surface of the catalyst, and the inside of the catalyst is never or scarcely used as sites for the reaction (col. 2, lines 34-38). The gas/liquid reactions that proceed in the presence of a water-repellent catalyst, such as an isotopic exchange reaction between water and hydrogen gas, require a gas and a liquid component to contact with the solid catalyst. Water cannot exchange its hydrogen atoms with hydrogen gas unless it is in contact with the solid catalyst. A water molecule must trap its hydrogen atom at a reaction site on the surface of the catalyst to get a substitute one trapped at another site in order to exchange them. The catalyst assembly of Mizumoto et al. ‘263 can be effectively utilized by letting the gas permeate the porous interior to reach the surface and then the reaction proceeds through the formation of three-phase interfaces.

In contrast, the present invention is directed to permeability of reactants and products through the porous catalyst layer. Then, the reaction proceeds as evidenced by the comparison of Examples 4 and 5, generating more by-products with a thicker catalyst (see Table 2 on page 43 of the present specification). The reason why a thicker catalyst layer produces worse selectivity of the tertiary amine is explained in the present specification. The present specification recites that “the process of transferring the reactants and the product in the inside of the catalyst is governed by diffusion, and the distance is reduced to 500 μm or less, whereby the mass transfer between the inside and outside of the catalyst can be promoted thereby effectively utilizing the whole of the catalyst and simultaneously suppressing the excessive reaction of the intermediate reaction product in the inside of the catalyst” (paragraph [0026] of the publication of the present application). If the reaction proceeded only on the surface of the film-type catalyst as on the surface of the catalyst assembly of Mizumoto et al. ‘263, the thickness of the film-type catalyst would not influence the reactivity.

This phenomenon is illustrated by comparing Example 1 and Comparative Example 1 (see Table 1 on page 42 of the present specification). The difference in the way the catalysts are prepared (which creates a difference in their internal structures) and the difference in thickness of the catalysts lead to very different results for the amination reaction. The film-type catalyst of Example 1 generates 95% of the desired amine with only 4% of undesired amine in five hours. In contrast, the pellet-type catalyst of Comparative Example 1, which has almost thirty times as much in weight as the film-type catalyst of Example 1, generates only 60% of the desired amine with 5% of the undesired amine. **As such, the film-type catalyst of the present invention can utilize the whole of the catalyst and simultaneously suppress the excessive reaction of the intermediate reaction product in the inside of the catalyst. This phenomenon is totally different from Mizumoto et al. ‘263 as described above.**

Turning to claims 3 and 13, the Examiner asserts that Mizumoto et al. ‘263 disclose a catalyst comprising copper. Applicants respectfully traverse. Mizumoto et al. ‘263 only disclose Pt, Ru, Rh, Ir, or an alloy thereof as a catalytically active noble metal (col. 3, lines 33-35).

Turning to claims 8 and 15, the Examiner relies on Mizumoto et al. '263 to disclose the subject matter of this claim. Applicants respectfully traverse. The substrate of Mizumoto et al. '263 is not a metal film. Rather, the substrate is a water-repellent porous polytetrafluoroethylene.

As discussed above, Mizumoto et al. '263 do not disclose each and every aspect of claims 1 and 10, from which all other claims ultimately depend. Applicants respectfully submit that Nishino et al. '355 and Yokota et al. '063 do not overcome the deficiencies of this reference.

To establish a *prima facie* case of obviousness of a claimed invention, all of the claim limitations must be disclosed by the cited references. As discussed above, the cited references fail to disclose all of the claim limitations of independent claims 1 and 10, and those claims dependent thereon. Accordingly, the combination of references does not render the present invention obvious.

Furthermore, the cited references or the knowledge in the art provide no reason or rationale that would allow one of ordinary skill in the art to arrive at the present invention as claimed. Therefore, a *prima facie* case of obviousness has not been established, and withdrawal of the outstanding rejections is respectfully requested. Any contentions of the USPTO to the contrary must be reconsidered at present.

Conclusion

All of the stated grounds of rejection have been properly traversed, accommodated, or rendered moot. Applicants therefore respectfully request that the Examiner reconsider all presently outstanding rejections and that they be withdrawn. It is believed that a full and complete response has been made to the outstanding Office Action, and as such, the present application is in condition for allowance.

Should there be any outstanding matters that need to be resolved in the present application, the Examiner is respectfully requested to contact Chad M. Rink, Registration No. 58,258, at the telephone number of the undersigned below to conduct an interview in an effort to expedite prosecution in connection with the present application.

If necessary, the Director is hereby authorized in this, concurrent, and future replies to charge any fees required during the pendency of the above-identified application or credit any overpayment to Deposit Account No. 02-2448.

Dated: November 19, 2010

Respectfully submitted,

By

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Exhibit I: R. B. Bird, et al., *Transport Phenomena*, John Wiley & Sons, Inc., New York, 1960 (pages 502-505).

Wiley International Edition

TRANSPORT PHENOMENA

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Preface

This book is intended to be an introduction to the field of transport phenomena for students of engineering and applied science. Herein we present the subjects of momentum transport (viscous flow), energy transport (heat conduction, convection, and radiation), and mass transport (diffusion). In this treatment the media in which the transport phenomena are occurring are regarded as continua, and very little is said about the molecular explanation of these processes. Surely the continuum approach is of more immediate interest to engineering students, although it should be emphasized that both approaches are needed for complete mastery of the subject.

Because of the current demand in engineering education to put more emphasis on understanding basic physical principles than on the blind use of empiricism, we feel there is a very definite need for a book of this kind. Obviously the subject matter is sufficiently basic that it cuts across traditional departmental lines. Our thought has been that the subject of transport phenomena should rank along with thermodynamics, mechanics, and electromagnetism as one of the key "engineering sciences." Knowledge of the basic laws of mass, momentum, and energy transport has certainly become important, if not indispensable, in engineering analysis. In addition, the material in this text may be of interest to some who are working in physical chemistry, soil physics, meteorology, and biology.

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Professors J. O. Hirschfelder and C. F. Curtiss of the University of Wisconsin, with whom we have had many years of pleasant association, first introduced our chemical engineering department to the subject of transport phenomena some ten years ago via a graduate course; our present course is in a sense a direct descendant of theirs.

Professor H. Kramers (Technische Hogeschool, Delft, Holland) in 1956 prepared a set of lecture notes entitled *Physisch Transportverschijnselen*, which represented the first attempt that we know of to teach transport phenomena to engineering students; one of us (R.B.B.) had the pleasure of spending a semester at Professor Kramers' laboratory as a Fulbright Lecturer and Guggenheim Fellow, during which period he profited very much from discussions related to the teaching of transport phenomena. Miss Jeanne O. Lippert deserves our warmest thanks for typing the bulk of the manuscript and some parts of it several times. We are deeply indebted to Mr. Stuart E. Schreiber for his tireless efforts in mimeographing and assembling the original set of notes. Also we wish to thank Miss Ellen Gunderson for her part in assisting us with the preparation of the manuscript.

R. B. B.
W. E. S.
E. N. L.

Contents

PART I. MOMENTUM TRANSPORT

Chapter 1	Viscosity and the Mechanism of Momentum Transport	3
*§1.1	Newton's Law of Viscosity	3
	*Example 1.1-1. Calculation of Momentum Flux	7
*§1.2	Non-Newtonian Fluids	10
*§1.3	Pressure and Temperature Dependence of Viscosity	15
	*Example 1.3-1. Estimation of Viscosity from Critical Properties	18
	*Example 1.3-2. Effect of Pressure on Gas Viscosity	19
§1.4	Theory of Viscosity of Gases at Low Density	19
	*Example 1.4-1. Computation of the Viscosity of a Gas at Low Density	25
	*Example 1.4-2. Prediction of the Viscosity of a Gas Mixture at Low Density	25
§1.5	Theory of Viscosity of Liquids	26
	*Example 1.5-1. Estimation of the Viscosity of a Pure Liquid	29
Chapter 2	Velocity Distributions in Laminar Flow	34
*§2.1	Shell Momentum Balances: Boundary Conditions	35
*§2.2	Flow of a Falling Film	37
	*Example 2.2-1. Calculation of Film Velocity	41
	*Example 2.2-2. Falling Film with Variable Viscosity	41

Contents	Contents	Contents	
<p>*§2.3 Flow through a Circular Tube</p> <p>*Example 2.3-1. Determination of Viscosity from Capillary Flow Data, 48</p> <p>Example 2.3-2. Bingham Flow in a Circular Tube, 48</p> <p>*§2.4 Flow through an Annulus</p> <p>§2.5 Adjacent Flow of Two Immiscible Fluids</p> <p>*§2.6 Creeping Flow Around a Solid Sphere</p> <p>*Example 2.6-1. Determination of Viscosity from Terminal Velocity of a Falling Sphere, 60</p>	<p>42</p> <p>133</p> <p>136</p> <p>138</p> <p>140</p> <p>140</p> <p>142</p>	<p>§4.3 Steady Two-Dimensional Potential Flow</p> <p>Example 4.3-1. Ideal Flow Around a Cylinder, 136</p> <p>Example 4.3-2. Flow into a Rectangular Channel, 138</p> <p>§4.4 Boundary-Layer Theory</p> <p>Example 4.4-1. Flow Near a Wall Suddenly Set in Motion, 140</p> <p>Example 4.4-2. Flow Near the Leading Edge of a Flat Plate, 142</p>	
<p>Chapter 3 The Equations of Change for Isothermal Systems</p> <p>§3.1 The Equation of Continuity</p> <p>§3.2 The Equation of Motion</p> <p>§3.3 The Equation of Mechanical Energy</p> <p>§3.4 The Equations of Change in Curvilinear Coordinates</p> <p>§3.5 Use of the Equations of Change to Set Up Steady Flow Problems</p> <p>*Example 3.5-1. Tangential Annular Flow of a Newtonian Fluid, 94</p> <p>*Example 3.5-2. Shape of the Surface of a Rotating Liquid, 96</p> <p>Example 3.5-3. Torque Relationships and Velocity Distribution in the Cone-and-Plate Viscometer, 98</p> <p>§3.6 The Equations of Change for Incompressible Non-Newtonian Flow</p> <p>*Example 3.6-1. Tangential Annular Flow of a Bingham Plastic, 104</p> <p>Example 3.6-2. Components of the Momentum Flux Tensor For Non-Newtonian Radial Flow between Two Parallel Disks, 106</p> <p>*§3.7 Dimensional Analysis of the Equations of Change</p> <p>*Example 3.7-1. Prediction of Vortex Depth in an Agitated Tank, 108</p>	<p>71</p> <p>74</p> <p>76</p> <p>81</p> <p>82</p> <p>92</p> <p>92</p> <p>96</p> <p>98</p> <p>101</p> <p>101</p> <p>104</p> <p>106</p> <p>107</p>	<p>153</p> <p>154</p> <p>154</p> <p>158</p> <p>160</p> <p>161</p> <p>162</p> <p>163</p> <p>165</p> <p>166</p> <p>166</p> <p>166</p> <p>166</p> <p>166</p>	<p>*§5.1 Fluctuations and Time-Smoothed Quantities</p> <p>*§5.2 Time-Smoothing of the Equations of Change for an Incompressible Fluid</p> <p>*§5.3 Semiempirical Expressions for the Reynolds Stresses</p> <p>*Example 5.3-1. Derivation of the Logarithmic Distribution Law for Tube Flow (Far from Wall), 161</p> <p>*Example 5.3-2. Velocity Distribution for Tube Flow (Near Wall), 163</p> <p>*Example 5.3-3. Relative Magnitude of Molecular and Eddy Viscosity, 165</p> <p>§5.4 The Second-Order Correlation Tensor and Its Propagation (the von Kármán-Howarth Equation)</p> <p>*Example 5.4-1. Decay of Turbulence Behind a Grid, 173</p>
<p>Chapter 6 Interphase Transport in Isothermal Systems</p> <p>§6.1 Definition of Friction Factors</p> <p>§6.2 Friction Factors for Flow in Tubes</p> <p>*Example 6.2-1. Pressure Drop Required for a Given Flow Rate, 188</p> <p>*Example 6.2-2. Flow Rate for a Given Pressure Drop, 189</p> <p>*§6.3 Friction Factors for Flow Around Spheres</p> <p>*Example 6.3-1. Determination of Diameter of a Falling Sphere, 194</p> <p>§6.4 Friction Factors for Packed Columns, 196</p>	<p>180</p> <p>181</p> <p>183</p> <p>188</p> <p>189</p> <p>190</p> <p>194</p> <p>196</p>	<p>Chapter 7 Macroscopic Balances for Isothermal Systems</p> <p>*§7.1 The Macroscopic Mass Balance</p> <p>*§7.2 The Macroscopic Momentum Balance</p> <p>*§7.3. The Macroscopic Mechanical Energy Balances (Bernoulli equation)</p> <p>Example 7.3-1. Derivation of Mechanical Energy Balance for Steady Incompressible Flow, 213</p>	
<p>*§4.1 Unsteady Viscous Flow</p> <p>*Example 4.1-1. Flow Near a Wall Suddenly Set in Motion, 124</p> <p>Example 4.1-2. Unsteady Laminar Flow in a Circular Tube, 126</p> <p>§4.2 Steady Viscous Flow With Two Nonvanishing Velocity Components: The Stream Function</p> <p>Example 4.2-1. "Creeping Flow" Around a Sphere, 132</p>	<p>123</p> <p>123</p> <p>124</p> <p>126</p> <p>130</p>	<p>208</p> <p>209</p> <p>210</p> <p>211</p> <p>214</p>	<p>*§7.4 Estimation of the Friction Loss</p> <p>*Example 7.4-1. Power Requirements for Pipe-Line Flow, 217</p>

Contents

*§7.5 Use of the Macroscopic Balances to Set Up Steady Flow Problems	219
*Example 7.5-1. Pressure Rise and Friction Loss in a Sudden Expansion, 219	
*Example 7.5-2. Performance of a Liquid-Liquid Ejector, 220	
*Example 7.5-3. Thrust on a Pipe Bend, 222	
*Example 7.5-4. Isothermal Flow of a Liquid through an Orifice, 224	
§7.6 Use of the Macroscopic Balances to Set Up Unsteady Flow Problems	226
Example 7.6-1. Efficacy Time for Flow from a Funnel, 226	
Example 7.6-2. Oscillation of a Damped Manometer, 229	

PART II ENERGY TRANSPORT**Chapter 8 Thermal Conductivity and the Mechanism of Energy Transport**

*§8.1 Fourier's Law of Heat Conduction	243
*Example 8.1-1. Measurement of Thermal Conductivity, 247	
*§8.2 Temperature and Pressure Dependence of Thermal Conductivity in Gases and Liquids	249
*Example 8.2-1. Effect of Pressure on Thermal Conductivity, 251	
§8.3 Theory of Thermal Conductivity of Gases at Low Density	253
Example 8.3-1. Computation of the Thermal Conductivity of a Monatomic Gas at Low Density, 258	
Example 8.3-2. Estimation of the Thermal Conductivity of a Polyatomic Gas at Low Density, 258	
Example 8.3-3. Prediction of the Thermal Conductivity of a Gas Mixture at Low Density, 259	
§8.4 Theory of Thermal Conductivity of Liquids	260
Example 8.4-1. Prediction of the Thermal Conductivity of a Liquid, 261	
§8.5 Thermal Conductivity of Solids	262

Chapter 9 Temperature Distributions in Solids and in Laminar Flow

*§9.1 Shell Energy Balances; Boundary Conditions	265
*§9.2 Heat Conduction with an Electrical Heat Source	266
*Example 9.2-1. Voltage Required for a Given Temperature Rise in a Wire Heated by an Electric Current, 271	
Example 9.2-2. Heating of an Electric Wire with Temperature-Dependent Electrical and Thermal Conductivity, 272	

Contents

*§7.5 Use of the Macroscopic Balances to Set Up Steady Flow Problems	219	Contents	xv
*Example 7.5-1. Pressure Rise and Friction Loss in a Sudden Expansion, 219		§9.3 Heat Conduction with a Nuclear Heat Source, 274	
*Example 7.5-2. Performance of a Liquid-Liquid Ejector, 220		*§9.4 Heat Conduction with a Viscous Heat Source, 276	
*Example 7.5-3. Thrust on a Pipe Bend, 222		§9.5 Heat Conduction with a Chemical Heat Source, 279	
*Example 7.5-4. Isothermal Flow of a Liquid through an Orifice, 224		§9.6 Heat Conduction through Composite Walls: Addition of Resistances, 283	
*Example 9.6-1. Composite Cylindrical Walls, 286		*Example 9.6-1. Composite Cylindrical Walls, 286	
*Example 9.7-1. Heat Conduction in a Cooling Fin		*Example 9.7-1. Error in Thermocouple Measurement, 290	
*Example 9.8-1. Forced Convection, 291		*§9.8 Forced Convection, 291	
*§9.9 Free Convection, 297		*§9.9 Free Convection, 297	
Chapter 10 The Equations of Change for Nonisothermal Systems 310			
*§10.1 The Equations of Energy	311	*§10.1 The Equations of Energy	
*§10.2 The Energy Equation in Curvilinear Coordinates	317	*§10.2 The Energy Equation in Curvilinear Coordinates	
*§10.3 The Equations of Motion for Forced and Free Convection in Nonisothermal Flow	317	*§10.3 The Equations of Motion for Forced and Free Convection in Nonisothermal Flow	
*§10.4 Summary of the Equations of Change	321	*§10.4 Summary of the Equations of Change	
*§10.5 Use of the Equations of Change to Set Up Steady-State Heat Transfer Problems	321	*§10.5 Use of the Equations of Change to Set Up Steady-State Heat Transfer Problems	
*Example 10.5-1. Tangential Flow in an Annulus with Viscous Heat Generation, 325		*Example 10.5-1. Tangential Flow in an Annulus with Viscous Heat Generation, 325	
*Example 10.5-2. Steady Flow of a Nonisothermal Film, 326		*Example 10.5-2. Steady Flow of a Nonisothermal Film, 326	
*Example 10.5-3. Transpiration Cooling, 328		*Example 10.5-3. Transpiration Cooling, 328	
*Example 10.5-4. Free-Convection Heat Transfer from a Vertical Plate, 330		*Example 10.5-4. Free-Convection Heat Transfer from a Vertical Plate, 330	
*Example 10.5-5. One-Dimensional Compressible Flow: Velocity, Temperature, and Pressure Gradients in a Stationary Shock Wave, 333		*Example 10.5-5. One-Dimensional Compressible Flow: Velocity, Temperature, and Pressure Gradients in a Stationary Shock Wave, 333	
*Example 10.5-6. Adiabatic Frictionless Processes in an Ideal Gas, 337		*Example 10.5-6. Adiabatic Frictionless Processes in an Ideal Gas, 337	
*§10.6 Dimensional Analysis of the Equations of Change	338	*§10.6 Dimensional Analysis of the Equations of Change	
*Example 10.6-1. Forced-Convection Heat Transfer in an Agitated Tank, 339		*Example 10.6-1. Forced-Convection Heat Transfer in an Agitated Tank, 339	
*Example 10.6-2. Surface Temperature of an Electric Heating Coil, 340		*Example 10.6-2. Surface Temperature of an Electric Heating Coil, 340	
Chapter 11 Temperature Distributions with More Than One Independent Variable 352			
*§11.1 Unsteady Heat Conduction in Solids	352	*§11.1 Unsteady Heat Conduction in Solids	
*Example 11.1-1. Heating of a Semi-Infinite Slab, 353		*Example 11.1-1. Heating of a Semi-Infinite Slab, 353	
*Example 11.1-2. Heating of a Finite Slab, 354		*Example 11.1-2. Heating of a Finite Slab, 354	
*Example 11.1-3. Cooling of a Sphere in Contact with a Well-Stirred Fluid, 357		*Example 11.1-3. Cooling of a Sphere in Contact with a Well-Stirred Fluid, 357	

Contents	Contents
<p>§11.2 Steady Heat Conduction in Laminar Flow of a Viscous Fluid</p> <p><i>Example 11.2-1: Laminar Pipe Flow with Constant Heat Flux at Wall</i>, 362</p> <p><i>Example 11.2-2: Laminar Pipe Flow with Constant Heat Flux at Wall: Asymptotic Solution for Small Distances</i>, 363</p> <p>§11.3 Steady Two-Dimensional Potential Flow of Heat in Solids</p> <p><i>Example 11.3-1: Temperature Distribution in a Wall</i>, 365</p> <p>§11.4 Boundary-Layer Theory</p> <p><i>Example 11.4-1: Heat Transfer in Forced-Convection Laminar Flow along a Heated Flat Plate</i>, 367</p>	<p>Chapter 14 Energy Transport by Radiation 426</p> <p>*§14.1 The Spectrum of Electromagnetic Radiation 427</p> <p>*§14.2 Absorption and Emission at Solid Surfaces 429</p> <p>*§14.3 Planck's Distribution Law, Wien's Displacement Law, and the Stefan-Boltzmann Law 433</p> <p>*Example 14.3-1. Temperature and Radiant-Energy Emission of the Sun, 437</p> <p>*§14.4 Direct Radiation between Black Bodies in Vacuo at Different Temperatures 437</p> <p>*Example 14.4-1. Estimation of the Solar Constant, 443</p> <p>*Example 14.4-2. Radiant Heat Transfer between Disks, 444</p> <p>*§14.5 Radiation between Nonblack Bodies at Different Temperatures 445</p> <p>*Example 14.5-1. Radiation Shields, 446</p> <p>*Example 14.5-2. Radiation and Free-Convection Heat Losses from a Horizontal Pipe, 448</p> <p>Example 14.5-3. Combined Radiation and Convection, 448</p> <p>§14.6 Radiant Energy Transport in Absorbing Media</p> <p>*Example 14.6-1. Absorption of a Monochromatic Radiant Beam, 451</p>
<p>Chapter 12 Temperature Distributions in Turbulent Flow 375</p> <p>*§12.1 Temperature Fluctuations and the Time-Smoothed Temperature 375</p> <p>*§12.2 Time-Smoothing the Energy Equation 377</p> <p>*§12.3 Semiempirical Expressions for the Turbulent Energy Flux</p> <p><i>Example 12.3-1. Temperature Profiles in Steady Turbulent Flow in Smooth Circular Tubes</i>, 380</p> <p>§12.4 The Double Temperature Correlation and Its Propagation:</p> <p>The Corrsin Equation</p> <p><i>Example 12.4-1. Decay Equation for the Double Temperature Correlation</i>, 386</p>	<p>Chapter 15 Macroscopic Balances for Nonisothermal Systems 456</p> <p>*§15.1 The Macroscopic Energy Balance 456</p> <p>*§15.2 The Macroscopic Mechanical Energy Balance (Bernoulli Equation) 460</p> <p>*§15.3 Summary of the Macroscopic Balances for Pure Fluids 462</p> <p>*§15.4 Use of the Macroscopic Balances for Solving Steady-State Problems</p> <p>*Example 15.4-1. The Cooling of an Ideal Gas, 463</p> <p>*Example 15.4-2. Parallel- or Counter-Flow Heat Exchangers, 465</p> <p>*Example 15.4-3. Power Requirements for Pumping a Compressible Fluid through a Long Pipe, 467</p> <p>Example 15.4-4. Mixing of Two Ideal-Gas Streams, 470</p> <p>*Example 15.4-5. Flow of Compressible Fluids through Head Meters, 471</p> <p>*§15.5 Use of the Macroscopic Balances for Solving Unsteady-State Problems</p> <p><i>Example 15.5-1. Heating of a Liquid in an Agitated Tank</i>, 473</p> <p><i>Example 15.5-2. Operation of a Simple Temperature Controller</i>, 476</p> <p><i>Example 15.5-3. Free Batch Expansion of a Compressible Fluid</i>, 480</p>
<p>§13.1 Definition of the Heat-Transfer Coefficient</p> <p>*Example 13.1-1. Calculation of Heat-Transfer Coefficients from Experimental Data, 394</p> <p>*§13.2 Heat-Transfer Coefficients for Forced Convection in Tubes</p> <p>*Example 13.2-1. Design of a Tubular Heater, 405</p> <p>*§13.3 Heat-Transfer Coefficients for Forced Convection around Submerged Objects</p> <p>*§13.4 Heat-Transfer Coefficients for Forced Convection through Packed Beds</p> <p>*§13.5 Heat-Transfer Coefficients for Free Convection</p> <p>*Example 13.5-1. Heat Loss by Free Convection from a Horizontal Pipe, 414</p> <p>*§13.6 Heat-Transfer Coefficients for Condensation of Pure Vapors on Solid Surfaces</p> <p><i>Example 13.6-1. Condensation of Steam on a Vertical Surface</i>, 418</p>	<p>411</p> <p>412</p> <p>415</p>

PART III MASS TRANSPORT Chapter 16 Diffusivity and the Mechanisms of Mass Transport *§16.1 Definitions of Concentrations, Velocities, and Mass Fluxes <i>Example 16.1-1. Relations among the Molar Fluxes</i> , 501 *§16.2 Fick's Law of Diffusion *§16.3 Temperature and Pressure Dependence of Mass Diffusivity <i>*Example 16.3-1. Estimation of Mass Diffusivity at Low Density</i> , 507 <i>*Example 16.3-2. Estimation of Mass Diffusivity at High Density</i> , 507 <i>Example 16.4-1. Computation of Mass Diffusivity at Low Density</i> , 512 *§16.5 Theories of Ordinary Diffusion in Liquids <i>Example 16.5-1. Estimation of Mass Diffusivity for a Binary Liquid Mixture</i> , 515 Chapter 17 Concentration Distributions in Solids and in Laminar Flow *§17.1 Shell Mass Balances: Boundary Conditions *§17.2 Diffusion Through a Stagnant Gas Film <i>*Example 17.2-1. Determination of Diffusivity</i> , 526 <i>Example 17.2-2. Diffusion Through a Nonisothermal Spherical Film</i> , 527 *§17.3 Diffusion with Heterogeneous Chemical Reaction <i>*Example 17.3-1. Diffusion with Slow Heterogeneous Reaction</i> , 531 *§17.4 Diffusion with Homogeneous Chemical Reaction <i>*Example 17.4-1. Gas Absorption with Chemical Reaction in an Agitated Tank</i> , 534 *§17.5 Diffusion into a Falling Liquid Film: Forced-Convection Mass Transfer <i>*Example 17.5-1. Gas Absorption from Rising Bubbles</i> , 541 *§17.6 Diffusion and Chemical Reaction Inside a Porous Catalyst: the "Effectiveness Factor" Chapter 18 The Equations of Change for Multicomponent Systems *§18.1 The Equations of Continuity for a Binary Mixture *§18.2 The Equation of Continuity of A in Curvilinear Coordinates §18.3 The Multicomponent Equations of Change in Terms of the Fluxes	Contents §18.4 The Multicomponent Fluxes in Terms of the Transport Properties 563 Chapter 18.5 Use of the Equations of Change to Set Up Diffusion Problems 572 <i>Example 18.5-1. Simultaneous Heat and Mass Transfer</i> , 572 <i>Example 18.5-2. Thermal Diffusion</i> , 574 <i>Example 18.5-3. Pressure Diffusion</i> , 575 <i>Example 18.5-4. Forced Diffusion</i> , 577 <i>Example 18.5-5. Three-Component Ordinary Diffusion with Heterogeneous Chemical Reaction</i> , 578 *§18.6 Dimensional Analysis of the Equations of Change for a Binary Isothermal Fluid Mixture <i>*Example 18.6-1. Blending of Miscible Fluids</i> , 582 Chapter 19 Concentration Distributions with More Than One Independent Variable 592 §19.1 Unsteady Diffusion <i>Example 19.1-1. Unsteady-State Evaporation</i> , 594 <i>Example 19.1-2. Unsteady Diffusion with First-Order Reaction</i> , 598 <i>Example 19.1-3. Gas Absorption with Rapid Chemical Reaction</i> , 599 §19.2 Boundary-Layer Theory: von Kármán Approximate Method <i>Example 19.2-1. Unsteady Evaporation into a Multicomponent Mixture</i> , 602 <i>Example 19.2-2. Diffusion and Chemical Reaction in Isothermal Laminar Flow Along a Soluble Flat Plate</i> , 605 §19.3 Boundary-Layer Theory: Exact Solutions for Simultaneous Heat, Mass, and Momentum Transfer <i>Example 19.3-1. Calculation of Mass-Transfer Rate</i> , 619 Chapter 20 Concentration Distributions in Turbulent Flow 626 <i>*§20.1 Concentration Fluctuations and the Time-Smoothed Concentration</i> 626 <i>*§20.2 Time-Smoothing of the Equation of Continuity of A</i> 627 <i>§20.3 Semiempirical Expressions for the Turbulent Mass Flux in Smooth Circular Tubes</i> , 630 <i>Example 20.3-1. Concentration Profiles in Turbulent Flow in Smooth Circular Tubes</i> , 630 <i>Example 20.3-2. Evaporation of Ammonia in a Wetted Wall Column</i> , 630 <i>§20.4 The Double Concentration Correlation and Its Propagation: the Corsin Equation</i> 633
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xx Chapter 21 *§21.1 Interphase Transport In Multicomponent Systems 636 *§21.2 Definition of Binary Mass-Transfer Coefficients in One Phase 637 Correlations of Binary Mass-Transfer Coefficients in One Phase at Low Mass-Transfer Rates 642 *Example 21.2-1. <i>Evaporation of a Freely Falling Drop</i> , 648 *Example 21.2-2. <i>The Wet-and-Dry-Bulb Psychrometer</i> , 649 *§21.3 Definition of Binary Mass-Transfer Coefficients in Two Phases at Low Mass-Transfer Rates 652 *§21.4 Definition of the Transfer Coefficients for High Mass-Transfer Rates 656 §21.5 Transfer Coefficients at High Mass-Transfer Rates: Film Theory 658 *Example 21.5-1. <i>Rapid Evaporation of a Pure Liquid</i> , 666 *Example 21.5-2. <i>Use of Correction Factors in Dropwise Evaporation</i> , 667 *Example 21.5-3. <i>Wet-Bulb Performance at High Mass-Transfer Rates</i> , 667 §21.6 Transfer Coefficients at High Mass-Transfer Rates: Penetration Theory 668 §21.7 Transfer Coefficients at High Mass-Transfer Rates: Boundary-Layer Theory 672 *Example 21.7-1. <i>Rapid Evaporation from a Plane Surface</i> , 676 §21.8 Transfer Coefficients in Multicomponent Systems Example 21.8-1. <i>Mass Transfer in a Fixed-Bed Catalytic Reactor</i> , 678	Contents Postface Appendix A Summary of Vector and Tensor Notation 715 §A.1 Vector Operations from a Geometrical Viewpoint 716 §A.2 Vector Operations from an Analytical Viewpoint 719 Example A.2-1. <i>Proof of a Vector Identity</i> , 722 §A.3 The Vector Differential Operations 723 §A.4 Second Order Tensors 726 Example A.4-1. <i>Proof of a Tensor Identity</i> , 731 §A.5 Integral Operations for Vectors and Tensors 731 §A.6 Vector and Tensor Components in Curvilinear Coordinates Example A.6-1. <i>Transformation Characteristics of Vector and Tensor Products</i> , 736 §A.7 Differential Operations in Curvilinear Coordinates Example A.7-1. <i>Derivation of Several Differential Operations in Cylindrical Coordinates</i> , 737	Contents Appendix B Tables for Prediction of Transport Properties 743 §B.1 Intermolecular Force Parameters and Critical Properties 744 §B.2 Functions for Prediction of Transport Properties of Gases at Low Densities 746	Contents Appendix C Constants and Conversion Factors 747 §C.1 Mathematical Constants §C.2 Physical Constants §C.3 Conversion Factors Notation 748 Author Index 747 Subject Index 748
Chapter 22 *§22.1 Macroscopic Balances for Multicomponent Systems 685 *§22.2 The Macroscopic Mass Balances 686 *§22.3 The Macroscopic Momentum Balance 688 *§22.4 The Macroscopic Mechanical Energy Balance 689 *§22.5 Use of the Macroscopic Balances to Solve Steady-State Problems Example 22.5-1. <i>Energy Balance for a Sulfur Dioxide Converter</i> , 690 *Example 22.5-2. <i>Height of a Packed-Tower Absorber</i> , 692 *Example 22.5-3. <i>Expansion of a Reactive Gas Mixture through a Frictionless Adiabatic Nozzle</i> , 697 §22.6 Use of the Macroscopic Balances for Solving Unsteady-State Problems Example 22.6-1. <i>Start-Up of a Chemical Reactor</i> , 700 Example 22.6-2. <i>Unsteady Operation of a Packed Column</i> , 702	Contents Appendix A Summary of Vector and Tensor Notation 715 §A.1 Vector Operations from a Geometrical Viewpoint 716 §A.2 Vector Operations from an Analytical Viewpoint 719 Example A.2-1. <i>Proof of a Vector Identity</i> , 722 §A.3 The Vector Differential Operations 723 §A.4 Second Order Tensors 726 Example A.4-1. <i>Proof of a Tensor Identity</i> , 731 §A.5 Integral Operations for Vectors and Tensors 731 §A.6 Vector and Tensor Components in Curvilinear Coordinates Example A.6-1. <i>Transformation Characteristics of Vector and Tensor Products</i> , 736 §A.7 Differential Operations in Curvilinear Coordinates Example A.7-1. <i>Derivation of Several Differential Operations in Cylindrical Coordinates</i> , 737	Contents Appendix B Tables for Prediction of Transport Properties 743 §B.1 Intermolecular Force Parameters and Critical Properties 744 §B.2 Functions for Prediction of Transport Properties of Gases at Low Densities 746	Contents Appendix C Constants and Conversion Factors 747 §C.1 Mathematical Constants §C.2 Physical Constants §C.3 Conversion Factors Notation 748 Author Index 747 Subject Index 748

§16.2 FICK'S LAW OF DIFFUSION

In Eq. 1.1-2 the viscosity μ is defined as the proportionality factor between momentum flux and velocity gradient (Newton's law of viscosity). In Eq. 8.1-6 the thermal conductivity k is defined as the proportionality factor between heat flux and temperature gradient (Fourier's law of heat conduction):

TABLE 16.2-1
EQUIVALENT FORMS OF FICK'S FIRST LAW OF BINARY DIFFUSION

Flux	Gradient	Form of Fick's First Law
n_A	$\nabla \omega_A$	$n_A - \omega_A(n_A + n_B) = -\rho \mathcal{D}_{AB} \nabla \omega_A$ (A)
N_A	∇x_A	$N_A - x_A(N_A + N_B) = -c \mathcal{D}_{AB} \nabla x_A$ (B)
j_A	$\nabla \omega_A$	$j_A = -\rho \mathcal{D}_{AB} \nabla \omega_A$ (C)
J_A^*	∇x_A	$J_A^* = -c \mathcal{D}_{AB} \nabla x_A$ (D)
j_A	∇x_A	$j_A = -\left(\frac{c^2}{\rho}\right) M_A M_B \mathcal{D}_{AB} \nabla x_A$ (E)
J_A^*	$\nabla \omega_A$	$J_A^* = -\left(\frac{c^2}{c M_A M_B}\right) \mathcal{D}_{AB} \nabla \omega_A$ (F)
$c(v_A - v_B)$	∇x_A	$c(v_A - v_B) = -\frac{c \mathcal{D}_{AB}}{x_A x_B} \nabla x_A$ (G)

Now we define the mass diffusivity $\mathcal{D}_{AB} = \mathcal{D}_{BA}$ in a binary system in an analogous fashion:

$$J_A^* = -c \mathcal{D}_{AB} \nabla x_A \quad (16.2-1)$$

This is *Fick's first law of diffusion*,¹ written in terms of the molar diffusion flux J_A^* . This equation states that species A diffuses (moves relative to the mixture) in the direction of decreasing mole fraction of A , just as heat flows by conduction in the direction of decreasing temperature.

A number of other mathematically equivalent statements of Fick's first law have appeared in the literature, and some of them are summarized in Table 16.2-1 for reference only. The diffusivity \mathcal{D}_{AB} is *identical* in all these equations. Of special importance in the following chapters is the form of Fick's first law in terms of N_A , the molar flux relative to stationary coordinates:

$$N_A = x_A(N_A + N_B) - c \mathcal{D}_{AB} \nabla x_A \quad (16.2-2)$$

This equation shows that the diffusion flux N_A relative to stationary coordinates is the *resistant* of two vector quantities: the vector $x_A(N_A + N_B)$,

¹ Temperature gradients, pressure gradients, and external forces also contribute to the diffusion flux, although their effects are usually minor. More complete expressions for the diffusion flux are given in §18.4.

which is the molar flux of A resulting from the bulk motion of the fluid, and the vector $J_A^* = -c \mathcal{D}_{AB} \nabla x_A$, which is the molar flux of A resulting from the diffusion superimposed on the bulk flow. Thus in Fig. 16.1-1b the bulk flow and diffusion terms in Eq. 16.2-2 are in the same direction for species A (because A is diffusing with the current) and are opposed for species B (because B is diffusing against the current). Note that the kinematic viscosity ν and the thermal diffusivity α also have the same units.

The way in which these three quantities are analogous can be seen from the following equations for the fluxes of mass, momentum, and energy in one-dimensional systems:

$$j_{Ay} = -\mathcal{D}_{AB} \frac{d}{dy} (p_A) \quad (\text{Fick's law for constant } \rho) \quad (16.2-3)$$

$$\tau_{yx} = -\nu \frac{d}{dy} (\rho v_x) \quad (\text{Newton's law for constant } \rho) \quad (16.2-4)$$

$$q_y = -\alpha \frac{d}{dy} (\rho \hat{C}_p T) \quad (\text{Fourier's law for constant } \rho \hat{C}_p) \quad (16.2-5)$$

These equations state, respectively, that (a) mass transport occurs because of a gradient in mass concentration, (b) momentum transport occurs because of a gradient in momentum concentration, and (c) energy transport occurs because of a gradient in energy concentration. These analogies do not apply in two- and three-dimensional problems, however, because τ is a tensor quantity with nine components, whereas j_A and q are vectors with three components.

In Tables 16.2-2, 3, and 4 some values of \mathcal{D}_{AB} are given for a few gases,

TABLE 16.2-2
EXPERIMENTAL DIFFUSIVITIES OF SOME DILUTE GAS PAIRS^a

Gas Pair	Temperature (°K)	\mathcal{D}_{AB} (cm ² sec ⁻¹)
CO ₂ —N ₂ O	273.2	0.096
CO ₂ —CO	273.2	0.139
CO ₂ —N ₂	273.2	0.144
	288.2	0.158
	298.2	0.165
Ar—O ₂	293.2	0.20
H ₂ —SF ₆	298.2	0.420
H ₂ —CH ₄	298.2	0.726

^a This table is abstracted from J. O. Hirschfelder, C. F. Curtiss, and R. B. Bird, *Molecular Theory of Gases and Liquids*, Wiley, New York (1954), p. 579. The values given are for 1 atm pressure.

TABLE 16.2-3
EXPERIMENTAL DIFFUSIVITIES IN THE LIQUID STATE^a

<i>A</i>	<i>B</i>	<i>T</i> (°C)	$\mathcal{D}_{AB} \times 10^6$ (cm ² sec ⁻¹)	x_A
Chlorobenzene	Bromobenzene	10.01	0.0332	1.007
			0.2642	1.069
			0.5122	1.146
			0.7617	1.226
			0.9652	1.291
		39.97	0.0332	1.384
			0.2642	1.691
			0.5122	1.806
			0.7617	1.902
			0.9652	1.996
Ethanol	Water	25	0.05	1.13
			0.275	0.41
			0.50	0.90
			0.70	1.40
		95	2.20	
Water	<i>n</i> -Butanol	30	0.131	1.24
			0.222	0.920
			0.358	0.560
			0.454	0.437
			0.524	0.267

^a This table is abstracted from a review article by P. A. Johnson and A. L. Babb, Liquid Diffusion in Non-Electrolytes, *Chem. Rev.*, **56**, 387-453 (1956); in this article a summary of experimental diffusion coefficients for liquid systems is given, as well as a survey of methods of measurement. Another excellent review article is that of L. J. Gosting, Measurement and Interpretation of Diffusion Coefficients of Proteins, *Advances in Protein Chemistry*, Vol. XI, Academic Press, New York (1956).

liquid, and solid systems. Diffusivities of gases at low density are almost composition independent, increase with the temperature, and vary inversely with pressure. Liquid and solid diffusivities are strongly concentration-dependent and generally increase with temperature. In the next three sections we summarize the available means for estimating diffusivities.

§16.3 TEMPERATURE AND PRESSURE DEPENDENCE OF MASS DIFFUSIVITY

The mass diffusivity \mathcal{D}_{AB} for a binary system is a function of temperature, pressure, and composition, whereas the viscosity μ and thermal conductivity

TABLE 16.2-4

EXPERIMENTAL DIFFUSIVITIES IN THE SOLID STATE^a

System	<i>T</i> (°C)	Diffusivity, \mathcal{D}_{AB} (cm ² sec ⁻¹)
He in SiO ₂	20	2.4 - 5.5 × 10 ⁻¹⁰
	20	4.5 × 10 ⁻¹¹
He in Pyrex	500	2 × 10 ⁻⁸
H ₂ in SiO ₂	500	0.6 - 2.1 × 10 ⁻⁸
		1.16 × 10 ⁻⁸
H ₂ in Ni	.85	
	165	10.5 × 10 ⁻⁸
Bi in Pb	20	1.1 × 10 ⁻¹⁶
	20	2.5 × 10 ⁻¹⁵
	20	3.5 × 10 ⁻²¹
	20	1.3 × 10 ⁻³⁰
	20	2.7 × 10 ⁻¹⁵
Hg in Pb	20	
Sb in Ag	20	
Al in Cu	20	
Cd in Cu	20	

^a Values taken from R. M. Barrer, *Diffusion in and through Solids*, Macmillan, New York (1941), pp. 141, 222, and 275.

k for a pure fluid are functions only of temperature and pressure. The data available on \mathcal{D}_{AB} for most binary mixtures are, moreover, quite limited in range and accuracy. The available correlations of \mathcal{D}_{AB} are of limited scope and are based more on theory than on experiment.

For binary gas mixtures at low pressure, \mathcal{D}_{AB} is inversely proportional to the pressure, increases with increasing temperature, and is almost independent of composition for a given gas-pair. The following equation for estimation of \mathcal{D}_{AB} at low pressures has been developed¹ from a combination of kinetic theory and corresponding-states arguments:

$$\frac{p\mathcal{D}_{AB}}{(p_A p_B)^{1/2} (T_A T_B)^{1/2}} = \left(\frac{1}{M_A} + \frac{1}{M_B} \right)^{1/2} = a \left(\frac{T}{T_A T_B} \right)^b \quad (16.3-1)$$

in which \mathcal{D}_{AB} [=] cm² sec⁻¹, p [=] atm, and T [=] °K. Analysis of experimental data gave the following values of the constants a and b :

For nonpolar gas-pairs:

$$a = 2.745 \times 10^{-4}$$

$$b = 1.823$$

For H_2O with a nonpolar gas:

$$a = 3.640 \times 10^{-4}$$

$$b = 2.334$$

¹ J. C. Slattery and R. B. Bird, *A.I.Ch.E. Journal*, **4**, 137-142 (1958).